Response to the reviewers' comments on "Radiative forcing of anthropogenic aerosols on cirrus clouds using a hybrid ice nucleation scheme"

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We thank editor and two reviewers very much for dedicating their time to read our manuscript and present important comments. We carefully studied these comments and revised the manuscript widely. Replies to these comments are listed point by point as below.

Reviewer #1

Summary:

The manuscript uses a newly developed ice nucleation scheme to examine the anthropogenic aerosol effect on cirrus clouds between preindustrial and present day conditions. The anthropogenic emissions have two opposite effects on the frequency of homogeneous ice nucleation and ice crystal number: soot particles decrease the homogeneous freezing and ice crystal number, while sulphur emissions enhance homogeneous nucleation. The total aerosol radiative effect is, moreover, strongly dependent on the freezing assumptions of the background state, as shown by the example of secondary organic aerosols. This manuscript has the potential to reveal some more details about the very uncertain anthropogenic aerosol effects on cirrus clouds. However, I am worried that the authors often describe features, which are not statistically significant. On one hand, they tried to avoid such feedbacks with limited success as the meteorological responses seem to often dominate over the cirrus microphysical responses to aerosols. I am therefore asking the authors to reassess their results after applying some form of a significance test. Adding several ensemble runs for each of their cases may help increasing the confidence in the presented results too. If those meteorological feedbacks turn out to still be important, than I would like to hear about them and understand them. This, together with my other comments, will demand substantial revision to the manuscript.

Major comments:

1.) I fear many of the conclusions of the paper at the regional level are not robust. The authors may be simply describing climatic noise and not climatic responses of cirrus cloud microphysics to changes in anthropogenic emissions, particularly what described on pages 15-19. In particular, I do not understand why the main signal in most of the panels in Figures 5,6,7,9,10,14 has the most pronounced anomaly between SE Asia and the W Pacific. What is causing that? We need to know it better, if that feedback is real/statistically significant? Would the same occur also in a longer (at least 10 years) free-running simulation? Considering the small changes due to the anthropogenic forcing, it may be valuable to run several ensembles for each of the cases.

Why is the meterological response so dominant if you are nudging your result?

On the other hand, why not allowing the full extent of the dynamical responses?

Ultimately, despite doing the nudging, the additional responses seem to often dominate over the pure aerosol signal.

It may also be beneficial to divide the radiative signal into the clear-sky and cloudy-sky (cloud radiative effect) components.

Re: The most pronounced differences in the sign of the impact of aerosols always occurs between SE Asia and the W Pacific because the number concentration of ice crystals is highest in that region as shown in Figure 2a. As a result, the perturbation of aerosols causes the largest absolute change there. We have added significance tests for the difference in Ni (Figure 3a, 4a, 5a, 11a), IWP (Figure 6a, 6c, 6e), LWP (Figure 6b, 6d, 6f) and radiative fluxes (Figure 7, 8, 9, 12) for all simulation differences and added a statement about the significance of the results in the paper. The radiative forcings in SE Asia due to the changes in sulfur emission (PD_Base-PI_SO4), and changes in all anthropogenic emission with and without SOA (PD_Base-PI_ALL and PD_SOA-PI_SOA) are statistically significant. However, the radiative forcing in W Pacific Ocean caused, in part, by meteorological feedback is not statistically significant. We have deleted the discussion on the forcing in W Pacific Ocean, but kept the discussion on the processes of meteorological feedback on the change in Ni. We have stated after the discussion as "Although these opposite changes in Ni due to meteorological feedbacks are not statistically significant (Figure 3a, 4a, 5a), our conjecture based on the model results indicates that the meteorological feedback caused by aerosol radiative effects might contribute to the change in Ni in remote regions. These important feedback processes need to be investigated further."

With small forcing like the forcing of aerosol on cirrus clouds it may be impossible to get a significant result without nudging. We tried to do a free running test with the same simulation in the past. However, we could not get a significant result after even a ten years free-running simulation. Chen and Gettelman (2013) found a much smaller radiative forcing from contrails cirrus (0.013±0.01 W m^{-2}) than our results (-0.14±0.07 W m^{-2}). They also tried to estimate the radiative forcing with a free running version of CAM5. Their study indicates that the detectable (95% confidence limit for Student's t test) globally averaged radiative flux perturbation at the top of the atmosphere to distinguish any radiative flux perturbation from model internal variability is 0.1 W m^{-2} from a 20 year simulation. The radiative forcing of aerosol on cirrus clouds is always near or lower than 0.1 W m^{-2} in our simulations, so the free-running mode of CAM5 is not an adequate modeling framework for estimating the radiative forcing on cirrus clouds. Chen and Gettelman (2013) indicate that a globally averaged perturbation above 0.01 W m^{-2} is statistically significant, based on the 95% confidence level of a Student's t test from a 20yr simulation using fixed meteorology.

Our model follows the nudging method in Zhang et al. (2014). Their study compares different nudging methods and indicates that compared to the wind-and-temperature nudging, constraining only winds leads to better agreement with the free-running model in terms of the estimated shortwave cloud forcing and the simulated convective activity. We only nudged the winds towards ECMWF reanalysis data in the model as stated in method section, so the meteorological responses are mostly due to the changes in temperature and humidity.

We have added a discussion of the clear sky radiative forcing (Lines 620, 636, 649, 731) and cloud radiative forcing for all cases and added the plots in Figure 7, 8, 9, 12.

Zhang, K., Wan, H., Liu, X., Ghan, S. J., Kooperman, G. J., Ma, P.-L., Rasch, P. J., Neubauer, D., and Lohmann, U.: On the use of nudging for aerosol–climate model intercomparison studies, Atmospheric Chemistry and Physics, 14, 8631-8645, 2014.

Chen, C.C. and Gettelman, A., 2013. Simulated radiative forcing from contrails and contrail cirrus. Atmos. Chem. Phys, 13(12), pp.525-12.

2.) The authors provide a few numbers of the estimated anthropogenic aerosol radiative forcing. However, the model used may allow you to further experiment with preindustrial (PI) and present day (PD) aerosol burdens/emissions in order to get a range of possible radiative forcings, given the large range of uncertainty and the inability to directly verify models with some form of observational data on the homogenous vs. heterogeneous freezing issue. Could you, for example, engineer your PI climate to produce an extreme low heterogeneous nucleation scenario and an extreme high heterogeneous nucleation scenario and apply at this point the anthropogenic aerosol forcing. You could also have two setups with the lowest and the highest plausible ice nucleating ability of soot.

To finish this comment on a positive note: I think your SOA experiment is a good example of some of those sensitivity experiments that I can foresee and would give us a better range of aerosol effects.

Re: We have added a sensitivity experiment (PD_HDust-PI_HDust) in the supplementary text S1 with the assumption of 100% of dust acting as INPs instead of only dust coating with fewer than 3 monolayers of sulfate acting as INPs in the base experiments (PD_Base-PI_ALL). The Ni from heterogeneous nucleation is larger by about a factor of ~4 in the PD_HDust compared to that in the PD_Base. With this extremely high heterogeneous nucleation, the all-sky net radiative forcing due to the changes in all anthropogenic emission from PI to PD in the sensitivity experiment is -0.16±0.06W m^{-2}, which is less negative than the radiative forcing of -0.20±0.05W m^{-2} in the base experiment. The results from the sensitivity experiment have been discussed in the supplementary text S1 and Figures S7-S9.

3.) The abstract reads as if a large part of the paper would be dedicated to a description of the newly developed scheme. However, this is a paper, which is using the scheme to estimate the aerosol effects on cirrus. You could include a more detailed description of your new freezing scheme.

Re: We have added the assumptions for aerosols to be effective INPs in the model in Table 1 and a detailed description of the new scheme in the method section which includes the method to calculate supersaturation, the treatment of gravity waves, as well as the calculation of ice number form heterogeneous nucleation and homogeneous nucleation. We added the statement "In the HYBRID scheme, the supersaturation (S_i) in the cloud parcel is calculated explicitly using the KL scheme so that ice particles are able to grow or decay throughout the time variations in the updrafts and downdrafts associated with gravity waves. S_i is calculated as a function of the updraft and aerosol concentrations at each grid. S_i is updated every second using

$$\frac{dS_i}{dt} = a_1 S_i w - (a_2 + a_3 S_i) \int_0^\infty dr_0 \frac{dn}{dr_0} R_{im}(r_0)$$

where the parameter a_1 is given by $a_1 = (L_s M_w g)/(c_p R T^2) - Mg/(RT)$, with the molar mass of air M and water M_w, latent heat of sublimation L_s, constant of gravity g, heat capacity at constant pressure c_p, the universal gas constant R, and air temperature T. w is the vertical velocity. a_2=1/n_{sat} with the water vapor number density at ice saturation n_{sat}. $a_3 =$

$L_s^2 M_w m_w/(c_p pTM)$, with the mass of a water molecule m_w and the air pressure p. R_{im} is the monodisperse freezing/growth integral,

$$R_{im} = \frac{4\pi}{v}\int_{-\infty}^{t} dt_0 \dot{n}_i(t_0) r_i^2(t_0,t)\frac{dr_i}{dt}(t_0,t),$$

where v is the specific volume of a water molecule. $dt_0\dot{n}_i(t_0)$ is the number density of aerosol particles that nucleate ice and freeze within the time interval between t_0 and $t_0 + dt_0$, $r_i(t_0,t)$ is the radius of the spherical ice particle at time t that froze and commenced to grow at time $t < t_0$, and dr_i/dt is the radial growth rate of that ice particle.

A series of updraft velocities at each grid point was generated based on a fitted wave spectrum of the observed equatorial gravity waves from Podglajen et al. (2016). The standard deviation of this wave spectrum was extended to other latitudes and seasons using the parameterization proposed by Gary (2008, 2006). It was extended vertically based on the static stability and atmospheric density. This parameterization of the wave spectrum associated with gravity waves is described in Penner et al. (2018).

When updraft velocity is positive, the LP parameterization is used to calculate the increase in the ice number from homogeneous and/or heterogeneous freezing, so that the HYBRID scheme avoids the lack of sensitivity to changes in aerosol number in the KL parameterization when calculating the number of new ice particles. The LP parameterization is derived by fitting the results of a large set of parcel model simulations covering different conditions in the upper troposphere (Liu and Penner, 2005). Two separate regimes are identified by the sign of $T - 6.07\ln w + 55.0$, where T is the temperature and w is the updraft velocity, to calculate the change of Ni due to homogeneous nucleation. When the sign is positive, the solution is in fast-growth regime for higher T and lower w. The number concentration of new ice crystals (N_i) is then calculated with the following equation

$$N_i = min\{exp(a_2)N_a^{a_1}exp(bT)w^c, N_a\}$$

where $b = b_1 lnN_a + b_2$, and $c = c_1 lnN_a + c_2$. N_a is the number concentration of sulfate in the Aiken and accumulation modes. The coefficients a_1, a_2, b_1, b_2, b_3, c_1, c_2 are constants, which can be found in Table 1 of Liu and Penner (2005). For lower T and higher w (the slow-growth regime), the following equation is applied to calculate N_i:

$$N_i = min\{exp[a_2 + (b_2 + b_3 lnw)T + c_2 lnw]N_a^{a_1+b_1T+c_1lnw}, N_a\}$$

where the coefficients $a_1, a_2, a_3, b_1, b_2, b_3, c_1, c_2$ are again listed in Table 1 of Liu and Penner (2005), and are different from those in the fast-growth regime. The number concentration of N_i from INPs in the heterogeneous nucleation regime is given as

$$N_i = min\{exp(a_{22})N_{INP}^{b_{22}}exp(bT)w^c, N_{INP}\}$$

where $b = (a_{11} + b_{11}lnN_{INP})lnw + (a_{12} + b_{12}lnN_{INP})$, and $c = a_{21} + b_{21}lnN_{INP}$. N_{INP} is the number concentration of total INPs. The coefficients $a_{11}, a_{12}, a_{21}, a_{22}, b_{11}, b_{12}, b_{21}, b_{22}$ can be found in the Section 4.2 of Liu and Penner (2005). When the updraft velocity is low and temperature is high, heterogeneous ice nucleation takes place initially and depletes the water vapor in the parcel so that homogeneous ice freezing never occurs. The threshold temperature T_c for heterogeneous nucleation-only is given by

$$T_c > aln(w) + b$$

where $a = -1.4938ln(N_{INP}) + 12.884; b = -10.41ln(N_{INP}) - 67.69$. When the regime is in a transition from heterogeneous-dominated to the homogeneous-dominated, the total ice number concentration from nucleation can be higher than the ice concentration from only heterogeneous nucleation, but lower than that from the pure homogeneous nucleation case. Then, N_i is interpolated from

$$N_i = N_{Het}\left(\frac{N_{Het}}{N_{Hom}}\right)^{\frac{N_{INP}-N_c}{0.9N_c}}$$

where N_{Het} is the ice number from pure heterogeneous nucleation, N_{Hom} is the ice number from pure homogeneous nucleation, N_{INP} is the number concentration of INPs and N_c is the critical number concentration of INPs for the heterogeneous nucleation-only regime."

Based on the method outlined above, HYBRID scheme calculates the increase in Ni using LP parameterization. The new ice crystals from nucleation either grow or decay with consumption/evaporation of water vapor and therefore change Si, which is determined using KL parameterization. The changes in Si then influence which particles are able to nucleate forming ice crystals.

It would be useful to have a more quantitative statement than "the ice number concentrations are in reasonable agreement/somewhat overestimated". Was there an improvement compared with the previous nucleation schemes used by the same group?

Re: We have added some quantitative evaluation as "The global model using the HYBRID scheme is able to do a reasonable job in predicting Ni with the difference in the median value between the simulation and observation less than 50% for all temperatures except for the high concentrations seen between 197K and 213K. The model predicts ~3 times higher *Ni* on average compared to the observations between 197K and 213K. Although the comparison of ice number concentration between our model and observation has not improved significantly compared to that shown by Penner et al. (2018), the new nucleation scheme improves the ability of nucleation to occur on small sized particles, since it avoids the calculation of ice nucleation chronologically from large sizes to small size used in the KL scheme, which results in an underestimation of ice crystals formed from small size particles." We have added this statement starting at Line 438.

I am also missing in particular a description of what aerosols species can nucleate ice heterogeneously and under what assumption. I understand the focus is on the soot, but the background loading of dust may have a huge impact on the magnitude of anthropogenic forcing. A sensitivity experiment with higher and another one with lower dust emissions may help addressing this issue.

Re: We have added Table 1 to describe the assumptions used for aerosols to be effective INPs in the model. Dust particles have an impact on the formation of ice crystals in the downwind regions of main dust sources (Sahara, Taklimakan) (Figure 2e). However, the impact is not very large, because we assumed only dust with fewer than 3 monolayers of sulfate coating are used to form heterogeneous INP in the model. This treatment is consistent with the results of field studies by *DeMott et al.* [2003], *Cziczo et al.* [2004] and *Richardson et al.* [2007]. We have stated this around Line 199. We have added a sensitivity experiment (PD_HDust-PI_HDust) in the supplementary text S1 with the assumption of 100% of dust acting as INPs instead of only dust coating with fewer than 3 monolayers of sulfate acting as INPs in the base experiments (PD_Base-PI_ALL). The *Ni* from heterogeneous nucleation is larger by about a factor of ~4 in the PD_HDust compared to that in the PD_Base with large concentrations in the downwind regions of dust sources (Figure S7). With this high heterogeneous nucleation case, the all-sky net radiative forcing due to the changes in all anthropogenic emissions from PI to PD in the sensitivity experiment is -0.16±0.06W m^{-2}, which is less negative than the radiative forcing of -0.20±0.05W m^{-2} in the base experiment. The results from the sensitivity experiment have been discussed in the supplementary text S1 and Figure S7-S9.

4.) What is your definition of a cirrus cloud? Is the paper referring only to the anthropogenic forcing on clouds at temperatures colder than the homogeneous freezing of water?

Re: We discussed the cirrus cloud formed from ice crystals in the paper. We define cirrus clouds for which the effects of aerosols are defined/calculated as all large-scale clouds formed at temperatures < -35°C. Cirrus clouds at these temperatures include anvil cirrus that are formed by the outflow from deep convection as well as large-scale cirrus formed by in-situ gravity waves. The detrained ice crystal number concentration in anvils is calculated from the detrained ice mass by assuming a spherical particle with a constant volume-mean radius, which is approximated as $3\rho_0 Q/(4\pi\rho_i r_{iv}^3)$ following Lohmann (2002). ρ_i is the ice crystal density, ρ_0 is the air density, riv is the volume mean radius determined from a temperature-dependent lookup table (Kristjánsson et al. 2000; Boville et al. 2006), Q is the detrainment rate of cloud water mass diagnosed from the convection parameterization. The new clouds generated by convective detrainment are assumed to be at saturation with respect to ice. In doing this, anvil clouds and in situ cirrus compete for the available water vapor within a grid box. When anvil clouds are formed due to convective detrainment, it reduces the saturation ratio in the clear-sky portion of a grid, and can potentially reduce the frequency of in situ large-scale cirrus formation (Wang and Penner 2010; Wang et al. 2014). We only calculate Ni as a result of the nucleation of aerosol in large-scale cirrus, so that when anvils occur in a grid box, we average the concentrations to determine the total ice number concentration in cirrus clouds. Anthropogenic emissions contribute to the change in the number concentration of ice crystals in large-scale cirrus cloud, but these are then averaged with the crystals in anvils. We have added this statement starting at Line 302.

Boville, B. A., P. J. Rasch, J. J. Hack, and J. R. McCaa, 2006: Representation of clouds and precipitation processes in the Community Atmosphere Model version 3 (CAM3). J. Climate, 19, 2184–2198.

Kristjánsson, J. E., J. M. Edwards, and D. L. Mitchell, 2000: Impact of a new scheme for optical properties of ice crystals on climates of two gems. J. Geophys. Res., 105, 10 063–10 079.

Lohmann, U., 2002: Possible aerosol effects on ice clouds via contact nucleation. J. Atmos. Sci., 59, 647–656.

Wang, M., Liu, X., Zhang, K., and Comstock, J. M., 2014: Aerosol effects on cirrus through ice nucleation in the Community Atmosphere Model CAM5 with a statistical cirrus scheme, Journal of Advances in Modeling Earth Systems, 6, 756-776, 10.1002/2014ms000339.

Wang, M., and Penner, J. E., 2010: Cirrus clouds in a global climate model with a statistical cirrus cloud scheme, Atmospheric Chemistry and Physics, 10, 5449-5474, 10.5194/acp-10-5449-2010.

Is the estimated forcing including the direct radiative forcing by increased aerosol burden? If so, please distinguish the clear sky effects from changes in cloud radiative effects.

Re: No, the direct radiative forcing by increased aerosol burden is not included in the forcing. The increased aerosol only influence ice nucleation in large scale cirrus clouds. We have added this statement around L246

5.)How does your model treat with cirrus with a origin at temperatures warmer than the homogeneous freezing threshold of water, also named "liquid-origin cirrus" (e.g. Krämer et al., 2016).

In particular, how are detrained ice crystals treated by the model? I understand the liquid-origin cirrus probably cannot be affected by aerosols, but the relative importance of detrainment vs. in-situ nucleation will substantially limit the potential for anthropogenic aerosol forcing in regions with frequent deep convection.

Re: please see response to comment 4.

6.)Please use a reasonable number of significant digits when providing results. Adding the third significant number likely makes no sense with only 6 years of simulations (e.g. page 7, line 163 and 164 and so on…).

Re: We have changed all numbers with three significant numbers to include two significant numbers.

7.)Some of the panels/figures need to be moved to the supplementary material due to the large amount of figures.

I also suggest that the vertical cross section plots are cut at about 700 hPa. Your focus is on cirrus clouds, while most of the plot area is wasted in the lower troposphere! (Fig. 3,4,5,6,13,14,15,16)

Re: We have moved 6 figures to supplementary material and cut the pressure axis at 700 hPa for all vertical plots in the paper.

Specific comments:

page 1, lines 17-19: please be more quantitative

Re: We have added the quantitative results here as "The ice number concentrations calculated using the HYBRID scheme (9.52±2.08 L^{-1}) are overestimated somewhat but are in reasonable agreement with those from the adiabatic parcel model (9.40±2.31 L^{-1})."

page 1, line 23 also lines 25 and 34:

The number of Ni doesn't mean much to the large majority of readers. Relative anomalies in units of % change may be more appropriate. Moreover, why do you focus on Ni only? A change in ice crystal radius will also contribute to changes in cirrus lifetime, leading to changes in cirrus optical properties.

Re: We have added the percentage of relative Ni anomalies in the abstract and main text. We focus on changes to Ni because a change in Ni depends directly on the change in aerosol number concentration. The radiative effects are frequently formulated in terms of effective ice crystal radius and ice water path, which together determine the optical depth. Effective ice crystal radius is calculated in the model by the ice water content and Ni. We focus on Ni and ice water path, since they are used in the model to determine effective radius, and are thus the primary variables affected by the change in aerosol number. The ice crystal radius is calculated by the number of Ni and ice water content in the model, so the change in radius is proportional to the change in the Ni to the 1/3 power if the ice water is constant.

page 2, lines 29-30:

Does this include both clear (direct radiative forcing) and cloudy sky changes? Please mention both.

Re: Yes, it is all-sky radiative forcing. We have added a discussion of the clear sky radiative forcing and cloud radiative forcing for all cases (Lines 620, 636, 649, 731) and added the plots in Figure 7, 8, 9, 12. Note that our clear sky radiative forcing is not associated with direct aerosol radiative forcing, but is rather due to changes in water vapor (see initial discussion by Wang and Penner, 2010)

page 2, line 47:

Considering that you are talking about changes to cirrus radiative effects, a reference like Hong et al., 2016 and/or Matus et al., 2017 may be useful.

Re: We have added these two references.

page 12, lines 283-285:

Could you also provide an effective or volumetric ice crystal radius histogram?

Re: The LP parameterization only calculates the number of ice nuclei but not the radius. The radius of ice crystals in the CESM model is calculated from the number of ice crystals and ice water content.

page 12, lines 305-307:

be more quantitative!

Re: We have changed this sentence to "The global model using the HYBRID scheme is able to do a reasonable job in predicting Ni with the difference in the median value between the simulation and observation less than 50% for all temperatures except for the high concentrations seen between 197K and 213K. The model predicts ~3 times higher *Ni* on average compared to the observations between 197K and 213K. Although the comparison of ice number concentration between our model and observation has not improved significantly compared to that shown by Penner et al. (2018), the new nucleation scheme improves the ability of nucleation to occur on small sized particles, since it avoids the calculation of ice nucleation chronologically from large sizes to small size used in the KL scheme, which results in an underestimation of ice crystals formed from small size particles."

page 13, lines 313-316:

What about dust? Shouldn't dust be the dominant source of ice nucleating particles at cirrus levels globally?

Re: Dust takes part in the heterogeneous nucleation, but we assume that only dust with fewer than 3 monolayers of sulfate coating are used to form heterogeneous INP in the model. When we assumed 100% of dust particles act as an ice nuclei in the sensitivity experiment (Text S1), the contribution to the INPs from dust are much larger (Figure S7). We understand that Cziczo et al. [2013] propose that dust are more important than other aerosols at cirrus levels based on their observations. However, these observations are primarily in regions where convection contributes mainly to the ice number through detrainment. Since anvil clouds are not included in our scheme for ice nucleation, dust can be a significantly smaller contributor to Ni.

Cziczo, D. J., Froyd, K. D., Hoose, C., Jensen, E. J., Diao, M., Zondlo, M. A., Smith, J. B., Twohy, C. H., and Murphy, D. M.: Clarifying the dominant sources and mechanisms of cirrus cloud formation, Science, 340, 1320-1324, 2013.

page 13, lines 328-330:

I do not understand why the changes are largest at about 200 hPa in a region, that should be deep convective detrainment, therefore coinciding with peak detrainment level. Is your model simulating a too low convective cloud top?

Alternatively, I could imagine that changes in homogeneous vs. heterogeneous freezing may not be as radiatively important in case the number of detrained ice crystals and ice water content dominates over the in- situ ice nucleation.

Re: As explained above (and now within the paper), we do not calculate the ice crystal number changes that result from convective detrainment. The changes are largest at about 200hPa because the emission of aircraft is around 200hPa.

Why is the effect not larger in the midlatitudes, where soot emissions are the largest?

Re: The number concentration of ice nuclei from homogeneous nucleation is largest in the tropics as shown in Figure 3(c), so the effect of inhibiting homogeneous nucleation as a result of adding the heterogeneous nucleation of soot is larger in the tropics although soot emission is larger in midlatitudes.

page 14, lines 353-354:

Direct effect vs. adjustments!

Re: The changes in the aerosol only have influence on the number concentration of ice nuclei which is indirect radiative effect on cirrus clouds. The direct radiative effect caused by the change of aerosols is not included. Clear sky radiative forcing in this study is not associated with direct aerosol radiative forcing, but is rather due to changes in water vapor which leads to changes in the clear sky longwave radiation (see initial discussion in Wang and Penner, 2010). We have added this statement in Line 421.

page 15, lines 366:

Why is there still a feedback on climatological state? Couldn't you use a longer simulation or nudge harder. A longer free-running simulation may tell something about the origin of those cloud adjustments, while a stronger nudging may prevent some of the noise to occur at first place.

Re: Please see the response to your first major comment.

page 15, lines 366- 394:

I am missing explanations that go beyond the "aerosol and cloud feedbacks to the meteorological state". Please describe what is really going on! Are the described patterns "real" at all? I am afraid that a lot of what mentioned is climatic or meteorologic "noise". Please apply a measure of statistical significance!

The meteorological feedbacks can be studied in an additional free running experiment. If such feedbacks are relatively speaking comparably or more important than the direct changes to cirrus freezing, we need to know more about them!

Re: As we discussed in Line 515, the changes in the aerosol and cirrus clouds lead to a change in the temperature. The changes in the temperature influence the homogenous nucleation. We have added the significance tests for the changes in

Ni (Figure 3a, 4a, 5a). The radiative forcing caused by meteorological feedback is not statistically significant. We have deleted the discussion of the forcing caused by the meteorological feedback, but keep the discussion on the processes of meteorological feedback on the change in Ni. We have stated after the discussion as "Although these opposite changes in Ni due to meteorological feedbacks are not statistically significant (Figure 3a, 4a, 5a), our conjecture based on the model results indicates that the meteorological feedback caused by aerosol radiative effects might contribute to the change in Ni in remote regions. These important feedback processes need to be investigated further."

Chapter 3.2:

I am a bit skeptical about the explanations of causes leading to changes in IWP, LWP, and radiative fluxes. How can you be sure you are seeing more than simple climatic variability? Adding a form of significance would be a first and easy step that would help clarifying this issue.

Re: We have added the significance tests for the difference in Ni, IWP, LWP and radiative fluxes in Figure 3, 4, 5, 6, 7, 8, 9, 12 and added the statement about the significance of the results in the paper. The explanations of causes are consistent with our understanding of the physics in the model, although it is very difficult to determine an explanation without many additional simulations trying to isolate process changes.

Moreover, if the meteorological adjustments play a larger role than the changes to cirrus clouds alone, you should dedicate part of your manuscript to those adjustments and try to understand them. In free-running experiments.

Please distinguish between changes to clear-sky and cloud radiative effects due to changes in emissions/freezing. Total net SW/LW/net radiative fluxes represent a mix of direct aerosol radiative effects and their impact on cirrus (and maybe also other cloud through meteorological feedbacks). Please show changes in clear sky and CRE separately!

Re: The direct aerosol radiative effects due to the change in anthropogenic emission are not included in the model. Instead, aerosol direct effects are only simulated in the CAM5 model using the CAM5 aerosol fields, which we do not change. Nevertheless, small changes in clear sky radiation could be caused by feedbacks to meteorology leading to changes in the CAM5 aerosol fields. The impact of aerosol on cirrus clouds and other clouds through feedbacks are the largest factors to change the radiation followed by changes in the clear sky water vapor, which leads to changes in the clear sky longwave radiation. We have added a discussion of the clear sky radiative forcing and cloud radiative forcing for all cases (Lines 620, 636, 649, 731) and added the plots in Figure 7, 8, 9, 12.

Moreover, it may be useful to separate the radiative perturbations on cirrus clouds only from the rest of the clouds. You could diagnose the cirrus cloud CRE with a help of a double call to the radiation routine, similarly to what is done for clear-sky radiative effects.

Re: The changes in IWP and LWP are shown in the Figure 6. We found the changes in LWP are all less than 0.5% with only a small number of significant grids (Line 583), so the influence of the change in warm clouds is not significant. The changes in aerosol only have a direct effect on the cirrus clouds in our model, while the changes in warm clouds are caused by feedbacks due to the change in cirrus clouds (Line 586). We have added the cloud radiative forcing for all cases and added a discussion of the cloud radiative forcing. The cloud forcing is mostly caused by the change in cirrus cloud.

page 18, lines 451-452:

Why is the FNT from soot largest around 30°N? Aren't the emissions much larger further north in the midlatitudes, around 40-60°N?

Re: As shown in Figure 3, the number of ice from homogeneous nucleation is large between 30°S and 30°N. The effect of soot is mainly caused by the suppression of homogeneous nucleation. So although the soot emission is larger around 40-60°N, the effect is larger around 30°N.

Fig. 3:

Why don't we see heterogeneous freezing that originates from the main dust sources (Sahara, Taklimakan, maybe Australia) in the panel e ? Is dust allowed to act as an ice nucleating particle?

Re: Dust is allowed to act as an ice nuclei in our model, but only about 1% of them are INP due to the restriction on having less than 3 monolayers of sulfate (see Table 1 in Penner et al., 2018). The ice crystal formed from dust can be found in the downwind region of main dust sources (Sahara, Taklimakan) in the Figure 2e, but the number concentration is not very large. The use of 3 monolayers to restrict the amount of dust that is an INP is consistent with the results of field studies by *DeMott et al.* [2003], *Cziczo et al.* [2004] and *Richardson et al.* [2007]. We have stated this around Line 198. In the text S1, when we assume 100% of dust act as INPs, the number concentration of INPs are much larger in the downwind region of dust sources (Figure S7).

Panels a,c,e miss units!

Re: We have added the units below the color bars.

Fig. 5,6,7,14,15,16:

Panels a,c,e miss units!

Re: We have added the units below the color bars.

Fig. 10,11,12,17 also miss units!

Re: We have added the units below the color bars.

Fig 10,11,12 and the corresponding text:

Please use a more descriptive naming for the FSNT,FLNT,FNT fluxes. Those abbreviation are not very intuitive to people outside of the CAM/CESM modeling community.

Re: We have changed them to shortwave radiation forcing (SRF), longwave radiative forcing (LRF) and net radiative forcing (NRF).

Reference:

- Hong and Liu, 2015: The Characteristics of Ice Cloud Properties Derived from CloudSat and CALIPSO Measurements, *JClim*
- Krämer et al., 2016: A microphysical guide to cirrus clouds – Part 1: cirrus types, *ACP*
- Matus and L'Ecuyer, 2017: The role of cloud phase in Earth's radiation budget, *JGR-A*

Reviewer #2:

In this work, Zhu and Penner implement a hybrid ice nucleation scheme in the CESM/IMPACT global climate model and perform simulations to quantify the impact of anthropogenic aerosol on cirrus clouds. The new scheme combines the best features of two existing cirrus parameterizations, in order to reduce their drawbacks and improve the resulting estimates of climate impacts.

The paper provides an important contribution to a research field that is still affected by large uncertainties and a relatively low level of scientific understanding. It also puts the results into the context of previous studies (although mostly citing works from the same group) and updates the estimates of the anthropogenic impacts on cirrus cloud properties.

There are, however, some parts of the paper that needs improvement, also for the sake of scientific reproducibility, and in general the presentation quality should be better structured and more accurate.

The comments given below should be addressed before the paper can be recommended for publication in ACP.

GENERAL COMMENTS:

1. The model description needs to be extended, as the limited amount of information provided may question the scientific reproducibility.

Re: We have added the assumptions for aerosols to be effective INPs in the model in Table 1 and a detailed description of the new scheme in the method section which includes the method to calculate supersaturation, determine the gravity wave spectrum, and calculate the ice number from heterogeneous nucleation and homogeneous nucleation. We added "In the HYBRID scheme, the supersaturation (S_i) in the cloud parcel is calculated explicitly using the KL scheme so that ice particles are able to grow or decay throughout the time variations in the updrafts and downdrafts associated with gravity waves. S_i is calculated as a function of the updraft and aerosol concentrations at each grid. S_i is updated every second using

$$\frac{dS_i}{dt} = a_1 S_i w - (a_2 + a_3 S_i) \int_0^\infty dr_0 \frac{dn}{dr_0} R_{im}(r_0)$$

where the parameter a_1 is given by $a_1 = (L_s M_w g)/(c_p R T^2) - Mg/(RT)$, with the molar mass of air M and water M_w, latent heat of sublimation L_s, constant of gravity g, heat capacity at constant pressure c_p, the universal gas constant R, and air temperature T. w is the vertical velocity. $a_2 = 1/n_{sat}$ with the water vapor number density at ice saturation n_{sat}. $a_3 = L_s^2 M_w m_w/(c_p p T M)$, with the mass of a water molecule m_w and the air pressure p. R_{im} is the monodisperse freezing/growth integral,

$$R_{im} = \frac{4\pi}{v} \int_{-\infty}^{t} dt_0 \dot{n}_i(t_0) r_i^2(t_0, t) \frac{dr_i}{dt}(t_0, t),$$

where v is the specific volume of a water molecule. $dt_0 \dot{n}_i(t_0)$ is the number density of aerosol particles that nucleate ice and freeze within the time interval between t_0 and $t_0 + dt_0$, $r_i(t_0, t)$ is the radius of the spherical ice particle at time t that froze and commenced to grow at time $t < t_0$, and dr_i/dt is the radial growth rate of that ice particle.

A series of updraft velocities at each grid point was generated based on a fitted wave spectrum to the observed equatorial gravity waves from Podglajen et al. (2016). The standard deviation of this wave spectrum was extended to other latitudes and seasons by using the parameterization proposed by (Gary, 2008, 2006). It was extended vertically based on the static stability and atmospheric density. This parameterization of the wave spectrum associated with gravity wave is described in Penner et al. (2018).

When updraft velocity is positive, the LP parameterization is used to calculate the increase in the ice number from homogeneous and/or heterogeneous freezing, so that the HYBRID scheme avoids the lack of sensitivity to changes in aerosol number in the KL parameterization when calculating the number of new ice particles. The LP parameterization is derived by fitting the results of a large set of parcel model simulations covering different conditions in the upper troposphere (Liu and Penner, 2005). Two separate regimes are identified by the sign of $T - 6.07 \ln w + 55.0$,where T is the temperature and w is the updraft velocity, to calculate the change of Ni due to homogeneous nucleation. When the sign is positive, it is fast-growth regime for higher T and lower . The number concentration of new ice crystals (N_i) is calculated with the following equation

$$N_i = min\{exp(a_2) N_a^{a_1} exp(bT) w^c, N_a\}$$

where $b = b_1 lnN_a + b_2$, and $c = c_1 lnN_a + c_2$. N_a is the number concentration of sulfate in Aiken and accumulation mode. The coefficients a_1, a_2, b_1, b_2, b_3, c_1, c_2 is constant, which can be found in Table 1 of Liu and Penner (2005). For lower T and higher w (the slow-growth regime), the following equation is applied to calculate N_i:

$$N_i = min\{exp[a_2 + (b_2 + b_3 lnw)T + c_2 lnw]N_a^{a_1+b_1T+c_1lnw}, N_a\}$$

where the coefficients a_1, a_2, a_3, b_1, b_2, b_3, c_1, c_2 are listed in Table 1 of Liu and Penner (2005), which are different with those in the fast-growth regime. The number concentration of N_i from INPs in the heterogeneous nucleation regime is given as

$$N_i = min\{exp(a_{22})N_{INP}^{b_{22}}exp(bT)w^c, N_{INP}\}$$

where $b = (a_{11} + b_{11} lnN_{INP})lnw + (a_{12} + b_{12} lnN_{INP})$, and $c = a_{21} + b_{21} lnN_{INP}$. N_{INP} is the number concentration of total INPs. The coefficients a_{11}, a_{12}, a_{21}, a_{22}, b_{11}, b_{12}, b_{21}, b_{22} can be found in the Section 4.2 of Liu and Penner (2005). When the updraft velocity is low and temperature is high, heterogeneous ice nucleation takes place initially and depletes the water vapor in the parcel so that homogeneous ice freezing never occurs. The threshold temperature T_c for the heterogeneous nucleation-only is given by

$$T_c > aln(w) + b$$

where $a = -1.4938ln(N_{INP}) + 12.884; b = -10.41ln(N_{INP}) - 67.69$. When the regime is transition from heterogeneous-dominated to the homogeneous-dominated, the total ice number concentration from nucleation can be higher than the ice concentration from heterogeneous nucleation, but lower than that from the pure homogeneous nucleation case. The N_i is interpolated from

$$N_i = N_{Het}\left(\frac{N_{Het}}{N_{Hom}}\right)^{\frac{N_{INP}-N_c}{0.9N_c}}$$

where N_{Het} is the ice number from pure heterogeneous nucleation, N_{Hom} is the ice number from pure homogeneous nucleation, N_{INP} is the number concentration of INPs and N_c is the critical number concentration of INPs for heterogeneous nucleation-only regime."

Based on the method outlined above, HYBRID scheme calculates the increase in Ni using LP parameterization. The new ice crystals from nucleation either grow or decay with consumption/evaporation of water vapor and therefore change Si, which is determined using

KL parameterization. The changes in Si then influence which particles are able to nucleate forming ice crystals.

2. Several types of INPs are considered by the HYBRID scheme, but their properties are only briefly mentioned in the text and it is hard to get an overview of what is assumed. Adding a table of all relevant INPs and their corresponding properties could be useful.

Re: We have added Table 1 to summarize the assumptions used in the model for all types of aerosol to be effective INPs in the model.

3. The results section is very hard to read. A lot of maps and panels are being mentioned, but not in the order they appear. Moreover some of the maps shown in the figures are not discussed. This part needs to be revised and restructured (like in different subsections). The number of figures could also be reduced, by moving the less relevant ones to a Supplement. The current presentation is overwhelming for the reader, who needs to browse through a large number of plots and maps (more than 80!), while only a very short text is given for each of them and no structured discussion is provided.

Re: We have moved 6 figures to a supplement and rearranged the figures. We have restructured paragraphs and tried to discuss the figures in order.

4. It is not clear whether the given changes in ice number concentration (Ni) and radiative forcing (RF) are statistically significant, as no statistical tests are applied or discussed. This is critically important for the difference map plots (Figs. 5-12 and 14-17), where some of the patterns depicted may be below the noise level.

Re: We have added significance tests for the difference in Ni, IWP, LWP and radiative fluxes in Figure 3, 4, 5, 6, 7, 8, 9, 12 and added a statement about the significance of the results in the paper. We have focused the discussion on the statistically significant aspects of radiative forcing.

5. All reported impacts on Ni from the various effects are given only as absolute values, while the relative changes would be useful to understand their relevance (especially for non experts).

Re: We have added the relative changes of Ni using percentage change in the abstract and main text.

SPECIFIC COMMENTS:

L23: is this value for Ni calculated over the whole column, or only at a specific altitude/temperature range?

Re: It is Ni over the whole column. We have added “over the entire column” to clarify what it is.

L23: here and in the rest of the manuscript, it would be useful to see the relative numbers (see also general comment 5).

Re: We have added the relative changes of Ni in percentage in the abstract and main text.

L27: this is quite low: did you check whether it is statistically significant?

Re: We have added the significance test in Figure 8 for the difference in radiative flux between PD_Base and PI_SO4. The net radiative forcing is statistically significant in south Asia, north Africa and north Indian Ocean. We have added a discussion on the significance of results in the results section.

L129: the turbulent kinetic energy as a proxy for subgrid scale vertical velocity was used in previous studies too, for example Lohmann et al. (J. Geophys. Res., 1999), Lohmann (J. Atmos. Sci, 2002), and Kärcher and Lohmann (J. Geophys. Res., 2002).

Re: We have added these references here.

L130: you could also mention Kuebbeler et al. (2014), who considered the contribution of orographic waves to the vertical velocity.

Re: We have stated "Kuebbeler et al. (2014) considered the contribution of orographic waves to the vertical velocity." around Line 134.

L169: please provide references for CESM and for IMPACT. L171: please specify the vertical resolution as well.

Re: We have stated CESM/IMPACT model has 30 vertical layers. The detail for CESM 1.2.2 can be found at http://www.cesm.ucar.edu/models/cesm1.2/. The IMPACT model refers to Liu et al. (2005) and more recent updates to IMPACT model that are introduced in the Methodology section.

L173: "fourteen species": which ones? L178: what is the hygroscopicity of bSoot?

Re: The fourteen species are introduced in the following, which are Soot (black carbon and organic carbon) from fossil fuel and biofuel burning, soot (black carbon and organic carbon) from biomass burning, aircraft soot activated within contrails and not activated, dust and sea salts in four size bins. The hygroscopicity of fSoot and bSoot is determined by volume averaging the hygroscopicity of the underlying particles and the number of sulfate monolayers on the particles. (Stated around Line 188).

L179: "with <1 monolayers of sulfate": does this mean no monolayers, hence purely hydrophobic soot? If yes, please rephrase.

Re: Not really, because the number of monolayers of sulfate is not an integer. <1 monolayers of sulfate means the particle is not covered by one monolayer of sulfate completely. We determine hygroscopicity by volume averaging the hygroscopicity of the underlying particle and the amount of sulfate.

L182–187: this sentence is unclear. It looks like there are 2 types of pre-activated soot. One freezes at 145% RHi, but it is not clear how the other type is treated in terms of ice nucleating ability.

Re: The other type of aircraft soot is not considered to act as INPs in our model. We have added this around Line 192.

L188–189: “Dust with fewer than 3 monolayers of sulfate coating is used to form heterogeneous INP in the model”. At which RHi?

Re: Dust may activate if the RHi reaches 120%. We have stated this in Table 1.

L206: are you considering the role of pre-existing ice crystal which may decrease the available supersaturation? The KL parameterization should include this possibility.

Re: While some schemes (e.g. Shi et al., ACP, 2015) consider that the initial nucleation in an updraft takes place in the presence of ice from the previous time step (i.e. “preexisting ice”, we do not. Rather, if our spectrum of waves leads to ice formation, and the next wave is an updraft, then the preexisting ice from the previous updraft is included in the KL scheme. If a wave within the spectrum considered in a GCM time step is a downdraft, then ice from the previous sub-time step may be evaporated if the depending on the downdraft velocity. We added “Unlike some schemes (e.g. Shi et al., 2015) which consider that the initial nucleation in an updraft takes place in the presence of ice from the previous time step, we assume the first parcel updraft within a GCM time step does not carry any preexisting ice, but thereafter if ice forms it may either grow and decrease the supersaturation or evaporate to some extent.”

L215–217: could you please be more specify on how this is technically realized in the model?

Re: We have added a detailed description of the method to calculate ice nucleation. See our respond to your general comment 1.

L226: in the CEDS dataset the historical series ends at 2014, so why not using a more recent year for PD instead of 2000? And what is meant by 2000? Is it the year 2000 or are the emissions varied transiently around 2000?

Re: Thanks for your suggestion. We will update the emission in our model to 2014 in the future, but we used the emission in year 2000 for this paper. We added that the year 2000 emissions are used for all six years of simulation.

L227: you may want to add that this is the same dataset used for the CMIP6 simulations.

Re: We have added "which are same with the emissions datasets used for the CMIP6 simulations"

L228–229: CEDS already provides aviation emissions, so apparently you are replacing them with the AEDT dataset in this work. Is there a reason for this choice? Is AEDT more accurate? Please elaborate on this, since emission data might be an important source of uncertainties in this kind of studies.

Re: The total BC emissions from aircraft in CEDS (5.8 Gg yr^{-1} in 2005 from Lee et al. (2009)) and the AEDT dataset (5.96 Gg yr^{-1}) for 2006 are very similar. We used the AEDT dataset in order to be able to evaluate the difference between our results in this paper with our previous results which all used the AEDT dataset. But perhaps more importantly, the original data on which AEDT emissions are based were developed based on the original flight tracks of each of the 31 million commercial flights worldwide (Wilkerson et al., 2010) and hence are presumed more accurate that those from Lee et al. (2009) which are based on IEA data for kerosene use by country. We have added this statement in Line 395.

Wilkerson, J. T., Jacobson, M. Z., Malwitz, A., Balasubramanian, S., Wayson, R., Fleming, G., Naiman, A. D., and Lele, S. K.: Analysis of emission data from global commercial aviation: 2004 and 2006, Atmos. Chem. Phys., 10, 6391–6408, https://doi.org/10.5194/acp-10-6391-2010, 2010.

Lee, D.S., G. Pitari, V. Grewe, K. Gierens, J.E. Penner, A. Petzold, M.J. Prather, U. Schumann, A. Bais, T. Berntsen, D. Iachetti, L.L. Lim and R. Sausen, 2010: Transport impacts on atmosphere and climate: Aviation, Atmos. Env., 44, (37), 4678 - 4734.

L231–233: I would suggest rephrasing this sentence as: "In a sensitivity experiment (PI_cSoot), the emission of cSoot...". Note that cSoot is not defined and it appears again later on in the manuscript.

Re: We have rephrased this sentence (Line 400) and defined cSoot around Line 192.

L240–242: which SOA precursors are considered? From which sources (natural/anthropogenic)? How do they change between PI and PD?

Re: The SOA used in the model is nucleated from highly oxygenated organic molecules (HOMs) formed from the oxidation of α-pinene and grown to accumulation mode size by sulfuric acid and oxidation products of isoprene, α-pinene, limonene and aromatics which partition to the aerosol phase. We have added this statement around Line 412. The differences in natural SOA precursors between PI and PD are caused by changes in temperature (which changes the isoprene, a-pinene and limonene emissions) as well as changes in land-use, while changes in aromatic emissions are associated with anthropogenic emission growth. Figure S6 of Zhu et al. (2019) show these differences.

Zhu, J., Penner, J. E., Yu, F., Sillman, S., Andreae, M. O., and Coe, H.: Decrease in radiative forcing by organic aerosol nucleation, climate, and land use change, Nature Communications, 10, 423, 10.1038/s41467-019-08407-7, 2019.

L246–248: are you using prescribed SSTs? Please specify.

Re: Yes, we have stated that the sea surface temperature is prescribed around Line 321.

L254: I would state here that this is a box model, as you are writing later in this section.

Re: We added it is a box model here.

L259–260: are you using this 2.2 min update frequency also when applying the scheme in the GCM? If yes, how do you realize that, given that the global model uses a 30-min time-step?

Re: Yes, the updraft velocity is updated every 2.2 min in the ice nucleation scheme in the CESM/IMPACT model. The values of updraft velocity are constant for a time interval of 2.2 minutes when simulating ice nucleation. The final interval within the 30 minute time step of the global model is shortened in order to match the 30 minute GCM time step. This final ice number concentration is passed back to the global model after this 30 minute interval. We have stated this around Line 337.

L302: for the sake of readability, you could consider splitting Sect. 3.1 in two parts: on the parcel model comparison (i.e., until L302) and on the GCM results (afterwards), respectively.

Re: We have split Section 3.1 in two parts.

L304: the Krämer dataset includes other interesting quantities, like ice water content and relative humidity. Why not comparing them as well?

Re: Because we updated the ice nucleation scheme which only output ice number to the global model and the changes in other quantities are caused by the change in the ice number, we focused on the evaluation of ice number.

L309: I would remove "somewhat" from this sentence: the simulated concentrations are about one order of magnitude higher than the observations around 205 K.

Re: We have deleted "somewhat".

L310–324: related to major comment 3 above: the figures should be discussed in the order they appear in the paper. Also some of the figures are not discussed at all (e.g., Figure 3d and 4d). In general, I find this paragraph quite hard to read and too short for the amount of the results that it should describe (two figures, with 5 lat-lon maps and 5 zonal plots). Please consider restructuring and expanding this part and the rest of the section.

Re: We have moved 6 figures to supplement and rearranged the figures. We have restructured paragraphs and tried best to discuss figures in order. The results are split into short paragraphs and expanded with the discussion on the clear sky radiative forcing and cloud forcing.

L311: the Ni spot over the tropics of Eastern Hemisphere in Figure 3a is remarkably high: could you please elaborate more about its possible causes?
Re: The large number concentration of sulfate in Aitken and accumulation mode in the upper troposphere (near 150hPa) over west Pacific Ocean and north Indian Ocean leads to the large Ni from homogeneous nucleation in the tropics of the Eastern Hemisphere. We have stated this around Line 452.

L323: how is the occurrence frequency calculated?
Re: The occurrence frequency of homogeneous nucleation is the ratio of the time steps when homogeneous nucleation occurs to all time steps. This is now defined (around line 510).

L325–339: same as for L310–324 and also related to major comment 3. L338: related to major comment 4, is this statistically significant?
Re: We have added a Student's t-test for the significance of the difference in Ni (Figure 3a). The change in Ni due to aircraft emission is significant in the North Atlantic Ocean and East coastal regions in North America (Line 480).

L341: "the increase in the sulfur emissions from PI to PD leads to a significant increase in Ni". Do you mean "statistically significant" or just "large"? If the first, a statistical analysis needs to be provided. For example, how much is the 90 or 95% confidence level on this quantity?
Re: We have changed to "large".

L380 and following: it is difficult to follow the discussion about these feedbacks without knowing how the model is set up. As I understood from Sect. 2, winds are nudged, but temperature is not. What about SST? How do you initialize the model in PI and PD? Please provide more details, either here or in Sect. 2.

Re: Sea surface temperature is prescribed using present day values for all cases. We have stated this around Line 425. However, feedbacks from the change in cirrus clouds can change the air temperature.

L385–387: could you please provide some numbers? Shipping is a large contributor to sulfur emissions over the oceans, and emission regulations in this sector have been introduced later than for other land-based sources (e.g., power plants).

Re: The global emission of sulfur in PI is 2.2 Tg S year^{-1}, while it is 55 Tg S year^{-1} in PD. The global emission of sulfur from shipping is 6.4 Tg S year^{-1}, which is much less than the emission of sulfur on land. We have added these numbers around Line 531.

L400–402: evaluating the model for IWC would be important for this discussion. See comment above about Figure 2: the Krämer dataset provides RHi and IWC, in addition to Ni. How are sedimentation of ice crystal and snow formation treated in the model? Are there differences between KL and LP? And if yes, how are they handled? I guess this is explained in the respective references, but it should be briefly mentioned in Sect. 2.

Re: We added the comparison between the global average IWP and CloudSat/CALIPSO analyses around Line 556. The global average IWP is 14.6 g m^{-2} in PD_Base case, which is lower than that observed in different CloudSat/CALIPSO analyses (21~28 g m^{-2}) (*Li et al.*, 2012). We used a cut off diameter of 250 μm to move cloud ice to snow. A cutoff diameter of 400 μm in the model almost doubles the IWP (compare IWP of dbfc_mg10 and dbfc in Table 3 in Penner et al., 2018). The KL and LP schemes only calculate ice nucleation, whereas the sedimentation of ice crystal and snow formation is calculated by the cloud microphysical processes in the

CESM. We have added this statement in Line 204.

L431: the acronym FSNT does not corresponds to what it describes (all-sky shortwave forcing).

L432: see previous comment.

L446–449: the acronyms FNT, FSNT, FLNT are not very useful, I would suggest replacing them with NET, SW, LW or something more intuitive.

Re: We have changed them to shortwave radiation forcing (SRF), longwave radiative forcing (LRF) and net radiative forcing (NRF)

L447–449: "The radiative forcing in cirrus clouds is mostly dominated by FLNT because of the larger longwave radiative effects of cirrus cloud than their shortwave radiative effects." This sounds like a circular argument: longwave radiative forcing dominates because longwave radiative effects are more important. I would suggest rephrasing this sentence.

Re: We have changed to a statement: The radiative effects in cirrus clouds are dominated by longwave radiative effects.

L476: what does the uncertainty range refer to? 1-sigma model variability? Or confidence level? Please specify. Statistical tests should also be performed.

Re: The uncertainty range is one standard deviation. We have specified with "(the uncertainty is the standard deviation of the interannual variation hereafter)" at the first time the uncertainty shown (around Line 121).

L532: see my comment at L341. L563: see my comment at L309.

Re: We have deleted "significant" or changed to "large" throughout the text.

Figure 2: please state that the red solid line shows the median (does it?) and also add a line for the median value of the observations.

Re: We have specified the red line is the median in the caption and added the median value of observations (blue dashed line) in Figure S1.

TECHNICAL CORRECTIONS:

L19: “observations” → “with observations”.

Re: We have added “with”.

L39: the acronym PI is already defined in the abstract.

Re: We have deleted “preindustrial period”.

L53: the acronym “GCMs” usually indicates “general” circulation models.

Re: We have deleted the acronym GCMs.

L106: “in limited studies” → “in a limited number of studies”.

Re: We have changed.

L114: “observations now indicate that”, this sounds like it is a new finding, but the fact that only a subsection of aerosols can act as INPs is well established (you also refer to studies from 2009, 2011 and 2012 to support this statement). I would rephrase this as "observations later indicated that” or similar.

Re: We have changed to “observations later indicated that”.

L163: please use a consistent number of decimal places for the RF results given throughout the paper. Even better would be to use mW m−2 instead of W m−2, given the small numbers involved (< 1 in absolute terms).

Re: We have changed all results to two significant numbers.

L163: there is a typo in the units (“W” is missing).

Re: We have added “W”.

L179: “with 1-3 monolayers” → “coated with 1-3 monolayers”.

Re: We have added “coated”.

L235: “(PI_ALL)” → “PI_ALL” (remove the brackets).

Re: We have removed the brackets.

L272–273: I would suggest using consistent units for the concentrations.

Re: We have changed 200 cm^{-3} to 0.2 L^{-1}.

L278 and L279: “with of the order” → “with concentrations of the order”.

Re: We have added “concentrations”.

L408: “in the Figure” → “in Figure”.

Re: We have deleted “the”.

L421: “IWP changes from” → “IWP switches from” (to avoid repetition).

Re: We have changed to “switches”.

L459: “the global average FNT due to sulfur emissions is a small negative, -0.025 0.064 W m−2”. This result is given as “-0.02 0.06 W m−2” in the abstract (L27). Please use a consistent number of decimal places for all numerical results in the paper.

Re: We have changed all results to include only two significant figures.

L565: “were used in the global model” → “were used in the CESM global model”.

Re: We have added “CESM/IMPACT”.

L573: “in the Table 2” → “in Table 2”.

Re: We have deleted “the”.

L577-580: please consider adding some punctuation in this long sentence.

Re: We have changed this sentence to "We found the possible effect of aerosol and cloud could feedback to the meteorological state such as temperature and RHi, which could have an opposite effect on the changes in Ni due to either aircraft soot or sulfur emissions in the remote regions like the west Pacific Ocean."

L582-585: see previous comment.

Re: We have changed this sentence to "The changes in Ni from PI to PD caused by all anthropogenic emissions are dominated by the changes due to the sulfur emissions, but the changes in surface and aircraft soot emissions have some effect on the inhibition of homogeneous nucleation."

L622: "observation" → "observations".

Re: We have changed.

L902: it looks like "900" does not belong here.

Re: We have deleted "900".